

DOCKET NO: 254914US0PCT

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF :
YASUO IMAMURA, ET AL. : EXAMINER: LIAO, DIANA J.
SERIAL NO: 10/500,737 :
FILED: JULY 6, 2004 : GROUP ART UNIT: 1793
FOR: HIGHLY PURE ULTRA-FINE :
SIOX POWDER AND ITS
PRODUCTION METHOD

APPEAL BRIEF

COMMISSIONER FOR PATENTS
ALEXANDRIA, VIRGINIA 22313

SIR:

This is an appeal under 35 U.S.C. § 134 to the Board of Patent Appeals and Interferences from the June 13, 2008, Final Rejection of Claims 4-8 and 10-15 of Application 10/500,737, filed July 6, 2004. The Notice of Appeal was timely filed on October 10, 2008.

STATEMENT OF REAL PARTY IN INTEREST

The real party in interest in this appeal is DENKI KAGAKU KOGYO KABUSHIKI KAISHA, having an address of 4-1, Yuraku-cho 1-chome, Chiyoda-ku, Tokyo 100-8455 Japan.

STATEMENT OF RELATED CASES

Appellants/Applicants, Appellants' legal representatives, and assignee(s) are aware of no appeals, interferences, or judicial proceedings that are related to, directly affect, or would be directly affected by, or have a bearing on the Board's decision in this appeal.

STATEMENT OF JURISDICTION

The Board of Patent Appeals and Interferences (Board) has jurisdiction over this appeal under 35 U.S.C. § 134. This is an appeal of a Final Rejection of pending Claims 4-8 and 10-15, dated June 13, 2008. A Notice of Appeal was timely filed on October 10, 2008. This Appeal Brief is timely filed on December 10, 2008, with no request for extension of time.

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STATUS OF AMENDMENTS

An amendment under 37 C.F.R. § 1.116, filed August 27, 2008, was entered of the record. The Advisory Action dated September 12, 2008, indicates that the amendment was entered and overcomes the objection to Claims 4-8 and 10-15.

GROUND S OF REJECTION TO BE REVIEWED

Claims 4-8 and 10-15 stand twice rejected under 35 U.S.C. § 103 as being unpatentable over Kumar et al. (U.S. Patent 6,726,990, issued April 27, 2004) in view of Bergonzo et al. (ELECTRONIC LETTERS; 31st March, 1994, Vol. 30, No. 7, pp 606-608).

STATEMENT OF FACTS

FACT

STATEMENT

Specification:

- I(1) The invention is directed to a method of producing SiO_x powder by reacting monosilane (SiH₄) gas with an oxidizing gas in a non-oxidizing gas atmosphere (Page 4, lines 19-23).
- I(2) The temperature of the reaction is in a range from 500 to 1000°C. (Page 5, line 6).
- I(3) The heat of the reaction is supplied by heating the periphery of the reactor (Page 6, lines 24-26; Page 9, lines 7-14).

- I(4) The oxidizing gas is not mixed with the monosilane prior to entering the reactor (Page 8, line 20, to Page 9, line 6; FIG. 1).
- I(5) The value of x in SiO_x is 0.6 to 1.8 (Page 7, lines 4- 12).
- I(6) Monosilane is superior to a halogen containing silane as a reactant because the halogen is not retained as a contaminant (Page 4, lines 20-23).
- I(7) A reaction temperature above 1000°C leads to the presence of impurities in the SiO_x powder (Page 5, lines 16-20).

Kumar et al. (U.S. 6,726,990):

- K(1) The silicon source is a halogenated or oxygenated silane (Kumar: Col. 3, lines 37-40).
- K(2) Reaction temperature is obtained by laser pyrolysis. Laser energy is absorbed by a radiation absorber included in the reaction mixture. The absorber absorbs the laser energy and transfers the energy to the reactants to drive the pyrolysis (Kumar: Col. 3, lines 53-56).
- K(3) Monosilane is listed as a radiation absorber (Kumar: Col. 3, line 52).
- K(4) Heating of the gases in the reaction zone is on the order of 10^5 degree C/sec (Kumar: Col. 5, lines 36-39).
- K(5) The reaction gases are mixed prior to entering the reaction chamber (Kumar: Col.4, lines 35-38).

- K(6) Laser pyrolysis is different from a thermal reaction (Kumar: Col. 3, lines 57-67).
- K(7) The heating rate in the laser pyrolysis reaction zone is on the order of 10^5 degree C/sec (Kumar: Col. 5, lines 36-39).
- K(8) Heat treatment of the laser pyrolysis formed particles is additionally required to obtain high purity particles (Kumar: Col. 7, lines 57-59; Col. 8, lines 42-49; Col. 9, lines 40-43).
- K(9) Silicon oxide nanoparticles for chemical-mechanical polishing are produced (Abstract).

Bergonzo et al. (ELECTRONIC LETTERS; 31st March, 1994, Vol. 30, No. 7, pp 606-608):

- B(1) A silicon dioxide (SiO_2) film is prepared by a photo deposition method (Bergonzo: page 607, Col. 1, lines 10-25).
- B(2) Reaction time is up to ten minutes (Bergonzo: page 607, Col. 1, lines 23-25).
- B(3) Substrate temperature during the deposition is a constant value between 100 and 500°C (Bergonzo: page 607, Col. 1, lines 17-18).
- B(4) The reaction mixture is composed of silane and oxygen and does not contain a radiation absorber (Bergonzo: page 607, Col. 1, lines 26-29).

ARGUMENT

Rejection of Claims 4-8 and 10-15 under 35 U.S.C. 103 over Kumar et al. (U.S. Patent 6,726,990) in view of Bergonzo et al. (ELECTRONIC LETTERS; 31st March, 1994, Vol. 30, No. 7).

1. Summary of the Invention

All claims stand or fall with independent Claim 4.

Claim 4 requires reaction of monosilane (SiH_4) gas with an oxidizing gas in a temperature range of 500 to 1000°C in a reactor which is heated to the reaction temperature by heat applied to the periphery of the reactor. The oxidizing gas is not mixed with the monosilane prior to entry to the reactor, but is mixed in the high temperature portion of the reactor. [Facts I(1-5)]

Surprisingly, Applicants have discovered that according to the claimed method, an ultra-fine, high purity SiO_x powder is directly obtained when monosilane is the silicon source and the reaction temperature is in the described temperature range [Facts I(6,7)].

2. Argument

The primary reference cited against the claims, Kumar, describes a method for producing silicon oxide nanoparticles involving laser pyrolysis and subsequently heating the particles produced by the laser pyrolysis under an oxidizing atmosphere at a temperature from about 400 to 800°C for a sufficient period of time to decolorize the particles [Facts K(2) and K(8)]. According to

the Kumar method, the silicon precursor is a compound with reasonable vapor pressure such as, for example, silicon tetrachloride, trichlorosilane, trichloromethylsilane and tetraethoxysilane [Fact K(1)].

The Examiner has cited this reference as disclosing a reaction temperature of 50-800°C (Official Action dated June 13, 2008, page 3, lines 10-11). However, the description cited is actually the treatment to obtain high purity particles [Fact K(8)] and not the reaction temperature as the Examiner erroneously finds. Applicants previous arguments did not directly address the Examiner's error, but described the actual reaction temperature as indicated in the following paragraph.

As Applicants argued in the Amendment and Request for Reconsideration, filed August 27, 2008 (page 6, line 14 to page 7, line 22), according to the Kumar laser method the combined gases are supplied in a well mixed state to a reaction chamber in a molecular stream [Fact K(5)]. A laser beam intersects the molecular stream and the radiation absorber absorbs the energy and distributes it to the reactants [Fact K(2)]. Kumar describes the reaction zone as follows (Col. 5, lines 36-43):

Heating of the reaction gases in reaction zone 224 is extremely rapid, roughly on the order of 10^5 degree C/sec depending on the specific conditions. The reaction is rapidly quenched upon leaving reaction zone 224, and particles 228 are formed in the molecular stream. The nonequilibrium nature of

the process allows for the production of nanoparticles with a highly uniform size distribution and structural homogeneity.

Kumar is silent with respect to residence time of the gas mixture in the reaction zone. However, according to this description, a residence time of one tenth of a second would lead to a reaction temperature of 1000°C and longer time would result in temperatures significantly higher.

The Examiner has erroneously asserted that heating of the reaction mixture via laser is equivalent to the thermal heating of the claimed invention (Official Action dated June 13, 2008, page 4, lines 3-9). However, Applicants herein submit that in contrast to the Examiner's opinion, Kumar states that the laser heating method is different [Fact K(6)] and describes the difference as follows:

Preferably, the energy absorbed from the radiation beam increases the temperature at a tremendous rate, many times the rate that energy generally would be produced even by strongly exothermic reactions under controlled condition. While the process generally involves nonequilibrium conditions, the temperature can be described approximately based on the energy in the absorbing region. The laser pyrolysis process is qualitatively different from the process in a combustion reactor

where an energy source initiates a reaction, but the reaction is driven by energy given off by an exothermic reaction.

Furthermore, Kumar describes mixing of the reaction gases prior to entrance of the reactor [Fact K(6)] whereas in the claimed invention the oxidizing gas is not mixed with the monosilane until entry to the high temperature portion of the reaction chamber [Fact I(4)].

The Office has acknowledged that Kumar is deficient in teaching SiH_4 as a Si source (Official Action dated June 13, 2008, page 6, lines 15-16).

Bergonzo is cited to show SiH_4 as a precursor gas.

Bergonzo describes a method for the photo-deposition of silicon dioxide (SiO_2) thin films using silane and oxygen mixtures [Fact B(1)]. This reference does not describe a partial oxide of formula SiO_x where x is 0.6 to 1.8 and it describes reaction times to form a thin film up to ten minutes [Fact B(2)]. Moreover, Bergonzo describes that the surface temperature of the substrate upon which the thin film is formed is between 100 and 500°C and is not increased by the lamp intensity [Fact B(3)].

Appellants have previously argued (Amendment and Request for Reconsideration, filed August 27, 2008, page 8, lines 1-7) that 1) Kumar is directed to production of particles of SiO_x ($1 \leq x \leq 2$), Bergonzo is directed to SiO_2 ; 2) Kumar is directed to reaction times measured in seconds or fractions of seconds, Bergonzo is directed to reaction times on the order of minutes; 3)

Kumar is directed to production of nanoparticles, Bergonzo is directed to producing a thin film; 4) Kumar requires an infrared absorber in the reaction gas mixture, Bergonzo does not; and finally 5) Kumar describes reaction conditions leading to temperatures of 1000°C and higher while Bergonzo describes a maximum temperature of 500°C.

The Examiner alleges that it would have been obvious to one of ordinary skill in the art to use SiH₄ as a silicon source in making silicon oxide in view of Bergonzo. (Official Action dated June 13, 2008, page 6, lines 19-20). However, as Appellants previously argued (Amendment and Request for Reconsideration, filed August 27, 2008, page 8, lines 8-21) in view of the above-cited significant differences in the reference descriptions, the Examiner has not explained how or why the teaching of the cited references prima facie would have led a person having ordinary skill in the art at the time of the invention to the subject matter claimed.

Moreover, Appellants note that Kumar describes SiH₄ as a radiation absorber [Fact K(3)]. Kumar is therefore aware of this material and does not disclose or suggest it as a silicon source for the laser pyrolysis process. Appellants urge that such lack of disclosure actually suggests that SiH₄ is not useful as a silicon source in the laser pyrolysis of Kumar.

In a Precedential Opinion rendered by the Board of Patent Appeals and Interferences (*Ex parte Whalen II*, Appeal 2007-4423, p. 16, lines 5-9, decided July 23, 2008) the Board stated:

The KSR Court [*KSR Int'l Co. v. Teleflex Inc.*, 550 U.S. 398 (2007)] noted that obviousness cannot be proven merely by showing that the elements of a claimed device were known in the prior art; it must be shown that those of ordinary skill in the art would have had some “apparent reason to combine the known elements in the fashion claimed.”

The Board further stated (*Id.* p.16, lines 17-25):

The Examiner has not persuasively explained why a person of ordinary skill in the art would have had a reason to modify the compositions taught by Evans, Greff'767, or Taki in a way that would result in the compositions defined by the claims on appeal. Therefore, The Examiner has not made out a prima facie case of obviousness under 35 U.S.C. § 103.

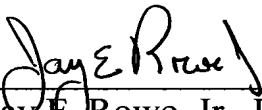
The Office has not reasonably explained how or why a person of ordinary skill in the art, at the time of the invention, would have combined the cited references to obtain the claimed invention. The Office has identified no motivation in either cited reference that would have led a person of ordinary skill in the art to the method of the claimed invention. Accordingly, the Office has not met its burden to show a prima facie case of obviousness.

Moreover, Kumar is directed to the production of silicon oxide nanoparticles for use in chemical-mechanical polishing [Fact K(9)], while Bergonzo is directed to a method for preparing a silicon dioxide film [Fact B(1)]. The two references do not pertain to the same field of endeavor, do not deal with the same problem, and are nonanalogous art. The Office has not explained why a person of ordinary skill in the art would have considered a reference dealing with the formation of a silicon dioxide thin film to solve the problem of forming silicon oxide nanoparticles. Accordingly, the Office has not met its burden to show a prima facie case of obviousness.

For all the above reasons, the rejection under 35 U.S.C. 103 as being unpatentable in view of Kumar et al. (U.S. Patent 6,726,990) and Bergonzo et al. (ELECTRONIC LETTERS; 31st March, 1994, Vol. 30, No. 7) should be reversed.

Respectfully submitted,

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APPENDIX

CLAIMS SECTION

Claim 1 (Withdrawn): A powder represented by the formula SiO_x wherein

x is from 0.6 to 1.8 and

the powder has a specific surface area of at least $10 \text{ m}^2/\text{g}$ and a total content of Na, Fe, Al and Cl of at most 10 ppm.

Claim 2 (Withdrawn): The powder according to Claim 1, wherein x is from 0.9 to 1.6.

Claim 3 (Withdrawn): The powder according to Claim 1, wherein the specific surface area is at least $50 \text{ m}^2/\text{g}$ and the total content of Na, Fe, Al and Cl is at most 5 ppm.

Claim 4 (Rejected): A method for producing a powder represented by the formula SiO_x comprising:

reacting monosilane gas with a gas capable of oxidizing the monosilane gas in a non-oxidizing gas atmosphere under a pressure of from 10 to 1000 kPa at a temperature of from 500 to 1000°C ,

wherein

x is from 0.6 to 1.8,

the SiO_x powder has a specific surface area of at least $10 \text{ m}^2/\text{g}$ and a total content of Na, Fe, Al and Cl of at most 10 ppm, and

in the reacting,

the gas capable of oxidizing the monosilane gas is supplied to a high temperature part of a reactor without prior mixing with the monosilane gas, and the temperature of 500 to 1000°C is obtained by heating the reactor on its periphery.

Claim 5 (Rejected): The method according to Claim 4, wherein the content in molar ratio of the non-oxidizing gas is at least twice the total amount of the monosilane gas and oxygen participating in the oxidation of the gas capable of oxidizing the monosilane gas.

Claim 6 (Rejected): The method according to Claim 4, wherein the gas capable of oxidizing the monosilane gas is oxygen, air, NO_2 , CO_2 or H_2O .

Claim 7 (Rejected): The method according to Claim 4, wherein the non-oxidizing gas is argon or helium.

Claim 8 (Rejected): The method according to Claim 4, wherein the pressure is from 50 to 300 kPa and the temperature is from 500 to 1000°C .

Claim 9 (Withdrawn): An interlayer dielectric film of a semiconductor device, a gas barrier film of a solar battery, a gas barrier film of a food packaging film or a protective film of an optical component, which comprises the SiO_x powder as defined in Claim 1.

Claim 10 (Rejected): The method according to Claim 4, wherein the reactor and gas introduction pipes are made of quartz.

Claim 11 (Rejected): The method according to Claim 4, wherein the value of x in the formula SiO_x is produced by changing the proportion of the monosilane gas to the oxidizing gas.

Claim 12 (Rejected): The method according to Claim 4, wherein the SiO_x powder is recovered by a powder recovery apparatus.

Claim 13 (Rejected): The method according to Claim 4, wherein the residual time of the monosilane gas and oxidizing gas in the reactor is from 0.2 to 1 second.

Claim 14 (Rejected): The method according to Claim 4, wherein the reacting is at a temperature of 550 to 950°C.

Claim 15 (Rejected): The method according to Claim 4, wherein the reacting is at a temperature of 650 to 850°C.

CLAIM SUPPORT AND DRAWING ANALYSIS SECTION

Claim 4 is the only independent claim involved in this appeal. In the following annotated claim the citations in braces provide specific support by reference to page and line number of the original specification.

Claim 4 (Rejected): A method for producing a powder represented by the formula SiO_x comprising:

reacting monosilane gas {**page 4, lines 19-20**} with a gas capable of oxidizing the monosilane gas {**page 4, lines 23-27**} in a non-oxidizing gas atmosphere {**page 5, line 25 to page 6, line 7**} under a pressure of from 10 to 1000 kPa at a temperature of from 500 to 1000°C {**page 5, lines 3-14**},

wherein

x is from 0.6 to 1.8 {**page 7, lines 7-10**},

the SiO_x powder has a specific surface area of at least 10 m²/g {**page 7, lines 18-20**} and a total content of Na, Fe, Al and Cl of at most 10 ppm {**page 7, lines 23-25**}, and

in the reacting,

the gas capable of oxidizing the monosilane gas is supplied to a high temperature part of a reactor without prior mixing with the monosilane gas {**page 9, lines 1-6, FIG. 1**}, and

the temperature of 500 to 1000°C is obtained by heating the reactor on its
periphery {**page 6, lines 24-26, page 9, lines 7-14, FIG. 1**}.

MEANS OR STEP PLUS FUNCTION ANALYSIS SECTION

There are no claims with means or step plus function language in the above-identified application.

EVIDENCE SECTION

Table of Contents

Item:

1. Kumar, S., et al., U.S. Patent No. 6,726,990, issued April 27, 2004, filed May 27, 1998.
2. Bergonzo, P., et al., ELECTRONIC LETTERS; 31st March, 1994, Vol. 30, No. 7, pp. 606-608.

Affidavits and Declarations

No affidavit or declaration is included in this Brief.

Other Evidence filed prior to the Notice of Appeal

None.

Other Evidence filed after the Notice of Appeal

Item:

3. *Whalen II, Ex parte*, Appeal 2007-4423, July 23, 2008.

RELATED CASES SECTION

Appellants/Applicants, Appellants' legal representatives, and assignee(s) are aware of no appeals, interferences, or judicial proceedings that are related to, directly affect, or would be directly affected by, or have a bearing on the Board's decision in this appeal.